

AMENDMENT UNDER 37 C.F.R. § 1.111

Application No.: 09/878,187

Atty Docket No.: Q61610

the claims over Harada et al. In the following discussion, applicants discuss each of the rejections and the Examiner's response to applicants' arguments concerning Harada et al.

Before discussing these rejections, applicants set forth the following Table that summarizes a comparison of the carbon fiber of the present invention, as exemplified by Example 1 of the present application, with the carbon fiber of Comparative Example 1 of the present specification and the disclosures of Harada et al, Tennent et al, Colomer and Lambert et al that have been cited by the Examiner.

Comparison of Present Invention with References

Table 1: Comparison of Carbon Fiber

Claim 13: A graphitized carbon fiber	Example 1	Comparative Example 1	Harada	Tennent	Colomer	Lambert
graphitized	○	○	○	○	×	×
discharging the impurity through a vicinity of highest-temperature section of the furnace while being accompanied by a carrier gas	○	×	×	×	×	×
the content of Fe, Ni or Co being 30 to 100 ppm	○	×	?	?	×	×

(Note) ○: agree ×: disagree ?: unknown

Claims 13 and 15 to 17 have been rejected under 35 U.S.C. § 102(b) as anticipated by/or, in the alternative, under 35 U.S.C. § 103(a) as obvious over Harada et al.

Applicants submit that Harada et al do not disclose or render obvious the presently claimed invention and, accordingly, request withdrawal of this rejection.

The carbon fiber of claim 13 of the present invention differs from conventional carbon fibers described at page 2 of the specification in that the carbon fiber of the present invention is, as described, for example, in Example 1, a carbon fiber having, for example, an Fe amount of 30 mass ppm (that is, about 100 ppm or less of a metal element) obtained after removing impurities from the gas outlet 25 positioned in the vicinity of the highest-temperature section in the furnace of an apparatus shown in Fig. 4, whereas conventional carbon fibers are a carbon fiber having an Fe amount of, for example, 200 ppm, as shown in the Comparative Example of the present specification, after removing impurities from the gas outlet 5 positioned at the low-temperature end in the furnace of the apparatus shown in Fig. 2.

The heat treatment temperatures that were employed in Example 1 and the Comparative Example were the same, namely, 2800°C. This difference in the carbon fibers seems to be attributable to the difference in the temperature distribution inside the heat-treatment furnace where the temperature is higher in the center portion as compared with the vicinity of the inlet of the heat-treatment furnace, and as a result, in the present invention impurities (metal components) can be efficiently discharged without being solidified. See page 9, lines 14 to 30 of the present specification.

Furthermore, in the removal of metal impurities according to the present invention, the percentage removal increases as the heat-treatment time (in the furnace) and the flow rate of carrier gas increase. Therefore, parameters such as heat treatment time and carrier gas flow rate may be determined by taking account of the required impurity standard for the vapor grown carbon fiber. See page 10, lines 2 to 6 of the present specification.

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Harada et al disclose performing a heat-treatment (graphitization) at 2000°C or more. However, Harada et al neither teach nor suggest how the impurities are removed, and how a vapor grown carbon fiber reduced in metal impurities can be obtained. By mere heat treatment at a high temperature, the amount of metal impurities does not decrease to 100 ppm or less, as proved in the Comparative Example of the present specification. See page 13, lines 5 to 6 of the present specification, which shows that a heat treatment at 2800°C in a conventional furnace, instead of the 2800°C heat treatment in Example 1 of the present specification, resulted in an Fe impurity amount of 200 ppm as compared to the 30 ppm impurity amount in Example 1.

The Examiner recognizes that Harada et al do not specifically disclose the metal impurity content of the graphitized fibers, but the Examiner asserts that “it is expected that it be within the claimed range of the applicant because no difference which would result in a graphitized fiber having more or less impurities is seen between the process of Harada et al and that of the instantly claimed invention”.

The Examiner further asserts that the process recitations of claim 15 do not further limit the claimed carbon fiber of independent claim 13.

In explaining his position, the Examiner states, at page 2 of the Office Action, that the only difference that he sees between Example 1 and Comparative Example 1 of the specification is the removal of the impurities from the inert gas, and that he does not see how the removal of impurities from a gas after its contact with the carbon fibers affects the carbon fibers themselves.

Harada et al disclose that the vapor grown fibers can be prepared by a vapor phase method, which can be substrate method or the fluidized growth method, both of which are

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disclosed by Harada et al as employing a metal catalyst. See column 4, line 51 to column 5, line 11. See also, column 1, lines 26 to 32, where Harada et al refer to an iron or nickel catalyst.

Harada et al do not provide any details on the subsequent heat treatment to bring about graphitization, except that the heat treatment is at a temperature of 2000°C or higher. In Example 1 of Harada et al, the temperature was 2800°C for 30 minutes in an argon atmosphere.

Harada et al do not disclose the concentration of metal in the vapor grown fiber prior to or subsequent to graphitization. Accordingly, applicants submit that it is impossible to provide a direct comparison based on Harada et al, since Harada et al do not disclose the amount of metal element. Applicants submit that the Comparative Example is an appropriate comparison, since Harada et al do not disclose the amount of catalyst in the vapor grown fiber, and an appropriate comparison, therefore, is one in which the starting amounts are the same, as in Example 1 and the Comparative Example of the present specification.

Applicants provide the following discussion of why Harada et al would achieve a high impurity content in the carbon fibers and why the removal of the gas from the highest-temperature section of the furnace affects the impurity content of carbon fibers themselves.

Applicants have argued that the difference in the carbon fibers between Example 1 and Comparative Example 1 seems to be attributable to the difference in the temperature distribution inside the heat-treatment furnace where the temperature is higher in the center portion as compared with the vicinity of the inlet of the heat-treatment furnace. Applicants provide the following further explanation of why the difference in the temperature distribution would result in a difference in the impurity content of the carbon fibers.

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In Harada et al, a vapor-grown carbon fiber obtained by pyrolysis is graphitized, but Harada et al only describe the use of an “argon atmosphere” during the graphitization and do not provide any details on whether and how the argon atmosphere is passed through and discharged from the furnace. Therefore, in Harada et al, a metal gas remaining in the atmosphere reattaches to the vapor-grown carbon fiber. In order to prevent this reattachment and obtain vapor-grown carbon fibers reduced in metal impurity, an atmosphere gas must be discharged as in the present invention, but Harada et al are silent on this. (See, the discussion in Harada et al under the heading “a Vapor-Grown and Graphitized Carbon Fibers”, beginning at column 3, lines 34.)

With respect to the method of Harada et al, only the following description is seen in Example 1 of Harada et al.

“Vapor grown carbon fibers having a mean fiber diameter of 0.8 μm , a mean fiber length of 132 μm and a mean aspect ratio of 165 were placed in an atmosphere in which argon gas is filled, and the carbon fibers were subjected to graphitization at 2,800°C for 30 minutes”.

From this description, one skilled in the art will generally consider as follows.

Since Harada et al only disclose that graphitization is performed in the state of argon gas being filled, a passing (discharge) of argon gas is not performed. Accordingly, the vaporized Fe as an impurity in Harada et al is not discharged but stays there, and as the temperature drops, Fe reattaches to VGCF.

In order to prevent reattachment, an atmosphere gas must be passed, but Harada et al ‘775 is silent on this. What is meant by “an atmosphere gas must be passed” is to merely “discharge” as shown in Fig. 1 which is the background art of the present invention or in Fig. 4

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which is the present invention. As described above, in the method of Harada et al, vaporized iron as an impurity stays and reattaches as the temperature drops and, therefore, its content becomes more than 100 ppm. Even in Comparative Example 1 which is a method of Fig. 1 disclosed as the background art of the present invention and in which argon gas is passed (discharged), 200 ppm of impurity is contained. Accordingly, it is apparent that if not passed, iron as an impurity is not discharged and its concentration is elevated. As a result, low impurity vapor grown carbon fibers having an iron content of 30 ppm as in Examples 1 and 2 of the present invention cannot be obtained.

In Example 1 of the present specification, where Fe does not reside in the graphitization furnace, VGCF having a very low Fe concentration can be stably obtained. In order not to allow the metal to reside in the furnace (in order to efficiently remove the metal), this can be suitably attained by discharging the carrier gas from the high-temperature section. As shown in Figs. 3 and 4 of the present application, the high-temperature portion is the center part of the furnace.

In Harada et al, pre-VGCF is continuously produced, but it cannot be thought that graphitization capable of removing the impurity Fe to the levels of the present invention is also performed by continuous operation.

Furthermore, the Fe amount in VGCF obtained by the method of Harada et al is not described. According to applicants' estimation, a large amount of Fe is contained. As described above, the Fe content in Comparative Example 1 of the present application, which is a method of decreasing the Fe amount more than in VGCF obtained by the method of Harada et al, is 200

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ppm, and it is theoretically apparent that the Fe amount in VGCF obtained by the method of Harada et al is larger than that.

In addition, applicants point out that a further difference between Example 1 and Comparative Example 1 is that in Example 1 there is less impurities in the final carbon fiber product than in Comparative Example 1. Thus, the Examiner's assertion that the only difference between the two examples is the removal of the impurities from the inert gas is not correct.

Further, in response to the Examiner's statement that he does not see how the removal of impurities from inert gas after its contact with the carbon fibers affects the carbon fibers themselves, applicants submit that it is not necessary for an applicant to understand the scientific reason why an improved result occurs, but that it is only necessary for an applicant to disclose how to achieve the improved result. However, applicants refer the Examiner to the above discussion.

The Examiner also argues that the Comparative Example in the specification does not prove that the high temperature heat-treatment performed other than by the method of Example 1 results in a metal content of greater than 100 ppm.

The Examiner states that the results shown in the Comparative Example are not persuasive because the Comparative Example discloses that the furnace in which the high temperature heating occurred was destroyed. The Examiner states that it "is expected that this not occur in the process of Harada et al, as a high temperature heating is of extreme importance to the overall teaching of Harada et al, and yet no mention is made to the destruction of the furnace" in Harada et al.

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The Examiner, therefore, concludes that the Comparative Example cannot be relied upon to determine the amount of metal impurity in the carbon fibers of Harada et al.

In response to this argument, and as stated in the Amendment Under 37 C.F.R. § 1.116 filed on March 22, 2004, Harada et al do not provide any details concerning the furnace and, therefore, no conclusions can be arrived at from Harada et al concerning whether or not there is a destruction of the furnace in Harada et al.

In further response to the Examiner's point that Harada et al do not discuss the destruction on the furnace, applicants submit the following.

When VGCF as much as 1,000 kg was processed as in Example 1 or in the Comparative Example, as shown in the Comparative Example, the argon was accompanied by impurity Fe, and as a result, Fe was deposited in the vicinity of furnace outlet where the temperature was low. Accordingly, the Fe concentration in the furnace increases, and even if the temperature is higher than the boiling point of Fe, VGCF having an Fe concentration of 100 ppm or less can be hardly obtained. In order to solve this problem, the impurity must be taken out from the high-temperature section of the furnace to the outside of the system by carrying it over with the argon gas.

Further, Harada et al do not disclose discharging of an inert gas at the graphitization.

Two reasons can be thought of why Harada et al do not make any mention of damage to the furnace.

One is because the impurity metal reattaches to the vapor-grown carbon fiber and does not transfer to the furnace.

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Another possible explanation is that high temperature heat-treatments are common in the prior art, and that it is a common occurrence for the furnace to be damaged and to be repaired or replaced before carrying out the next run of heat-treatment. Since it is a common occurrence for damage and subsequent repair or replacement to occur, one of ordinary skill in the art would not expect Harada et al to discuss this common occurrence. Since Harada et al do not make any mention of damage to the furnace or repair or replacement, one of ordinary skill in the art would expect that Harada et al experienced damage as commonly occurs, and that Harada et al would repair or replace the furnace before the next heat-treatment run as is common in the art.

The Examiner also argues that Harada et al teach the treatment of vapor grown carbon fibers identical to those of the applicant under equivalent temperatures “using the same carrier gas as the applicant”. The Examiner states that where the claimed products and the prior art products are identical or substantially identical or are produced by identical or substantially identical processes, the burden of proof is shifted to the applicant to prove that the prior art products do not necessarily or inherently possess the characteristics of the claimed product.

Applicants submit that they have satisfied this burden by the showing in Comparative Example 1. Since the only difference between Example 1 and Comparative Example 1 was the heat-treatment furnace where the temperature was higher in the center portion as compared with the vicinity of the inlet of the heat treatment furnace, applicants submit that this comparison proves that the mere use of identical heat-treatment temperatures does not produce the same result if the furnace conditions differ from each other.

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Since Harada et al do not provide any disclosure on the specific furnace they employ, and do not disclose the concentration of metal in the vapor grown carbon fibers prior or subsequent to graphitization, it is impossible to provide a direct comparison based on Harada et al.

Comparative Example 1, however, provides an appropriate comparison that can be made in the present circumstances, and disproves the Examiner's position that identical vapor grown carbon fibers will be obtained when using equivalent temperatures and the same carrier gas.

Further, Harada et al are silent on the passing or discharging of an inert gas at the graphitization. The mere fact that Harada et al may be "using the same carrier gas" does not provide any information on how that gas is used at the time of graphitization. The simple fact is that Harada et al do not disclose a discharging of the argon gas. If the argon gas in Harada et al is not discharged from the high-temperature section of the furnace as in the present invention, the impurity Fe is accumulated in the furnace, as discussed above.

In view of the above, applicants submit that Harada et al do not defeat the patentability of the present claims and, accordingly, request withdrawal of this rejection.

Claims 13 and 15 to 17 have been rejected under 35 U.S.C. § 103(a) as obvious over Tennent et al in view of the newly cited Lambert et al publication.

Applicants submit that Tennent et al and Lambert et al do not disclose or render obvious the presently claimed invention and, accordingly, request withdrawal of this rejection.

Tennent et al do not set forth any details on the method of the heat treatment. Tennent et al do disclose that a metal catalyst particle is employed. Therefore, since Tennent et al disclose a metal catalyst, applicants submit that it would be expected that the fiber filament disclosed at

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column 2 would contain metal impurity, and that since Tennent et al do not disclose the specific heat treatment set forth in the present claims, the fiber filaments described in Tennent et al would be expected to have a metal content of more than 100 ppm, such as illustrated in the Comparative Example of the present specification.

The Examiner recognizes that Tennent et al do not specifically teach a metal catalyst impurity content between 30 and 100 ppm, but relies on Lambert et al for teaching a process in which “most of the catalyst” was removed by a heat-treatment.

The Examiner argues that it would have been obvious to apply the Lambert et al treatment to the fibers of Tennent et al in order to obtain a purified product.

The Examiner asserts that it “is expected that the resulting product will have remaining catalyst metal impurities in an amount 30 and 100 ppm because no difference is seen between the process of Lambert et al and that of the instantly claimed invention”. The Examiner states that the Lambert et al method comprises heating the samples under vacuum at high temperatures, for example, 1600°C.

In response, Lambert et al disclose that carbon nanotubes can be produced either as multi-shell nanotubes (also known as “multiwalled nanotubes” or MWNT”) or as single-shell nanotubes (also known as “single-walled nanotubes” or “SWNT”). Lambert et al disclose that single-shell nanotubes are produced by using an electric arc-discharge process that employs a metallic catalyst. The Lambert et al publication is particularly directed to a process for producing high purity single shell nanotubes by eliminating the remaining catalyst particles in the final product, as disclosed at page 364, second column.

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Thus, in Lambert et al, the carbon nanotube was produced by carbon sublimation by an electric arc-discharge process. As disclosed in Lambert et al, a carbon nanotube produced by an arc process is a single walled nanotube (SWNT). This SWNT differs from a graphite structure such as obtained in the present invention.

A graphite structure of the present invention is a laminate structure of graphen sheets. The SWNT of Lambert et al, however, consists of one graphen sheet layer, and does not satisfy the definition of a graphite structure.

The studies by Lambert et al employed an arc process. In the arc process, the temperature of the reaction field is high as compared with a thermal decomposition method, which is the method recited in the present claims. The thermal decomposition method produces vapor grow carbon fiber (VGCF) that has a multiwalled structure. In the case of MWNT, its graphite structure is readily grown and a clear image is observed through a transmission electron microscope (TEM).

The production process of VGCF by a “thermal decomposition method” as in the present claims is a process of obtaining VGCF by “thermally decomposing” hydrocarbon, which is a carbon source of MWNT. On the other hand, the production method of SWNT in Lambert et al is an “arc process” of obtaining carbon nanotubes (CNT) by sublimating carbon. Thus, the production method of SWNT in Lambert et al is not a thermal decomposition method. The treatment of impurities obtained by the Lambert et al electric arc-discharge method is excluded in the present invention.

Further, in Section 3.3, beginning at page 366, Lambert et al describe their attempts to purify nanotubes by vacuum annealing at high temperature. In Section 3.3, the carbon nanotubes were made by use of a cobalt catalyst.

In Section 3.3, at page 366, right-hand column, Lambert et al state:

Fig. 3a shows a portion of the sample annealed for one day, revealing that most of the cobalt has been removed from the sample. Apparently, carbon nanotubes are not modified by this heat-treatment, i.e. they are not opened. There is still some carbon soot in the sample and some modified carbon structure have appeared. First, we can see numerous graphite shells that are left behind in samples after the catalyst particles have disappeared. It seems that the remaining cobalt particles have been trapped in these graphite shells. For a short annealing time (Fig. 3b), these new carbon shells are quickly formed with most of them containing small cobalt particles. (Emphasis added).

In the Discussion of Section 4, at page 370, right-hand column, Lambert et al state that:

...the catalyst particles could not be completely removed (traces remained as seen from Figs. 3a and 3b) even after a few days of annealing and this could mean that the catalyst has reacted to form a carbonaceous species with a high vapor pressure such as, for example, metal carbides. In the best case, the most efficient purification method leads to more than 90% of the catalyst particles being removed. (Emphasis added).

Since Lambert et al state that the most efficient purification method leads to more than 90% of the catalyst particles being removed, applicants submit that about 10% of the catalyst particles would remain. Applying this 10% figure to the 2% mass of catalyst particles employed in Example 1 of the present application, one would expect that there would be 2000 ppm of catalyst particles that would remain if the Lambert et al treatment were applied to Example 1.

Further, applicants submit that one of ordinary skill in the art would not be led to employing the Lambert et al method with the vapor grown particles of Tennent et al.

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Claims 13 and 15 to 17 have been rejected under 35 U.S.C. § 103(a) as obvious over Tennent et al taken with the newly cited Colomer et al publication.

Applicants submit that Tennent et al and Colomer et al do not disclose or render obvious the subject matter of claims 13 and 15 to 17 and, accordingly, request withdrawal of this rejection.

Applicants have discussed Tennent et al in detail above, and rely on that discussion.

In this rejection, the Examiner relies on the Colomer et al publication for a teaching of a method of removing catalyst material that remain in vapor grown carbon nanotubes.

The Colomer et al method comprises stirring a nanotube sample into a solution of hydrofluoric acid to dissolve the metal particle.

In Section 3.1, Colomer et al state that the separation of the nanotubes from the catalyst was carried out using hydrofluoric acid to dissolve the zeolitic support and the metal (catalyst) particles. This treatment was applied with stirring for 24 hours, followed by filtering and washing with distilled water. Colomer et al state that this allows the “total elimination of catalyst (confirmed by the disappearance of the characteristic X-ray peaks on the powder to fraction pattern spectra).”

In Colomer et al, Co supported as a catalyst on zeolite was reacted at 600°C by using acetylene as the carbon source, and zeolite and Co (catalyst) were dissolved in HF. Colomer et al state that since the corresponding peak disappeared as confirmed by powder X-ray diffraction, the catalyst was eliminated. However, the quantitative limit in the powder X-ray diffraction is

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The methods of Tennent et al and the present invention all are a thermal decomposition method and the production conditions differ from those of Lambert et al. The method for forming a SWNT in Lambert et al is a carbon sublimation method and cannot be characterized as being a method for forming a vapor grown carbon fiber by a thermal decomposition method.

Applicants submit that one of ordinary skill in the art would not be led to combining the teachings of Tennent et al with Lambert et al with Lambert et al because Tennent et al relate to multiwalled carbon nanotubes (MWNT), whereas the teachings of Lambert et al relating to removal of catalyst particles are directed to single wall carbon nanotubes (SWNT).

In addition, the reduction in the amount of catalyst particles disclosed in Lambert et al occurs in SWNT, which is formed by a different process than the thermal decomposition method disclosed in Tennent et al. There is no disclosure or suggestion in Lambert et al that a similar reduction in the amount of catalyst particles could be achieved in a MWNT formed by a thermal decomposition.

Further, although the Examiner states that he sees no difference between the process of Lambert et al and that of the presently claimed invention, Lambert et al do not disclose a high temperature heat treatment in which a carrier gas is discharged from the highest temperature section of the furnace. In addition, Lambert et al do not disclose the temperature recited in claim 15.

In view of the above, applicants submit that the combination of Tennent et al with Lambert et al would not have led one of ordinary skill in the art to the present invention and, accordingly, request withdrawal of this rejection.

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approximately 0.5 wt% (5,000 ppm). Accordingly, even if the peak disappeared, this does not mean that the impurity content was from 30 to 100ppm.

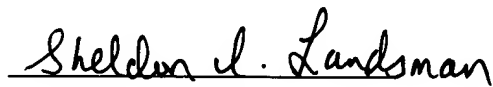
Accordingly, applicants submit that the present invention can not be achieved by combining the graphitization of Tennent et al and the dissolution in HF of Colomer et al.

In view of the above, applicants submit that Tennent et al and Colomer et al do not defeat the patentability of claims 13 and 15 to 17 and, accordingly, request withdrawal of this rejection.

In view of the above, reconsideration and allowance of this application are now believed to be in order, and such actions are hereby solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

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